



MAIL STOP APPEAL
BRIEF - PATENTS

IN THE UNITED STATES PATENT AND TRADEMARK OFFICE

Applicants: J.E. Sealey II et al. Attorney Docket No. WEYE117898/23441B

Application No.: 09/975,670 Group Art Unit: 1731

Filed: October 10, 2001 Examiner: M.S. Alvo

Title: PROCESS FOR MAKING A COMPOSITION FOR CONVERSION TO
LYOCELL FIBER FROM AN ALKALINE PULP HAVING LOW
AVERAGE DEGREE OF POLYMERIZATION VALUES

TRANSMITTAL OF APPEAL BRIEF

Seattle, Washington 98101

February 11, 2005

TO THE COMMISSIONER FOR PATENTS:

This Appeal Brief is being filed in response to the Notification of Non-Compliant Appeal Brief, mailed on January 11, 2005.

The Notification states the prior Appeal Brief is defective because:

1. The Brief improperly has a separate heading for Claims 7 and 8 under the 35 U.S.C. 112 rejection. The Examiner states the claims were not separately rejected under 35 U.S.C. 112.
2. The Brief does not contain a separate heading for each ground of rejection as required by 41.37(c)(1)(vii). The Examiner states Claims 1-11 are rejected under 35 U.S.C. 103(a) as being unpatentable over Gannon et al. (6,042,769) in view of WO 99/16960 and Samuelsson et al. (5,985,097); and alternatively Claims 1-11 are rejected under 35 U.S.C. 103(a) as being unpatentable over Gannon et al. (6,042,769) in view of WO 99/16960 and Samuelsson et al. (5,985,097) and further in view of Luo et al. (6,210,801).

Appellants respectfully call attention to 37 C.F.R. § 41.37(c)(i)(vii) that requires "[f]or each ground of rejection applying to two or more claims, the claims may be argued separately or as a group ... Any claim argued separately should be placed under a subheading identifying the claim by number." Therefore, Appellants believe that a separate heading for Claims 3, 7, 8, and 11 is proper, regardless of whether the claim is rejected under Section 103(a) or 112.

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Appellants have provided a separate heading for the rejection of the grouped Claims 1-11 under 35 U.S.C. 103(a) as being unpatentable over Gannon et al. (6,042,769) in view of WO 99/16960 and Samuelsson et al. (5,985,097) without Luo et al. (6,210,801).

The Commissioner is hereby authorized to charge any fees under 37 C.F.R. §§ 1.16, 1.17 and 1.18 which may be required during the entire pendency of the application, or credit any overpayment, to Deposit Account No. 03-1740. This authorization also hereby includes a request for any extensions of time of the appropriate length required upon the filing of any reply during the entire prosecution of this application. A copy of this sheet is enclosed.

Respectfully submitted,

CHRISTENSEN O'CONNOR
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I hereby certify that this correspondence is being deposited with the U.S. Postal Service in a sealed envelope as first class mail with postage thereon fully prepaid and addressed to Mail Stop Appeal Brief - Patents, Commissioner for Patents, P.O. Box 1450, Alexandria, VA 22313-1450, on the below date.

Date: February 11, 2005



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IN THE UNITED STATES PATENT AND TRADEMARK OFFICE
BEFORE THE BOARD OF PATENT APPEALS AND INTERFERENCES

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Application No: 09/975,670 Group Art Unit: 1731
Filed: October 10, 2001 Examiner: M.S. Alvo
Title: PROCESS FOR MAKING A COMPOSITION FOR CONVERSION TO
LYOCELL FIBER FROM AN ALKALINE PULP HAVING LOW
AVERAGE DEGREE OF POLYMERIZATION VALUES

APPELLANTS' APPEAL BRIEF

Seattle, Washington

February 11, 2005

TO THE COMMISSIONER FOR PATENTS:

This brief is filed in response to the Notification of Non-Compliant Appeal Brief, mailed on January 11, 2005. This brief was originally mailed on October 6, 2004, in support of a Notice of Appeal filed in the above-identified application on July 6, 2004, to the Board of Patent Appeals and Interferences appealing the decision dated March 9, 2004, of the primary Examiner finally rejecting Claims 1-11.

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I. REAL PARTY-IN-INTEREST

The real party-in-interest in the above-identified application is the assignee, Weyerhaeuser Company, a Washington corporation, having a place of business at 33663 Weyerhaeuser Way South, Federal Way, Washington.

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II. RELATED APPEALS AND INTERFERENCES

None.

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III. STATUS OF CLAIMS

Claims 1-11 are pending in the application. Claims 1-11 are appealed, and all stand rejected under 35 U.S.C. § 103(a) and 35 U.S.C. § 112, second paragraph. A copy of the claims on appeal, as currently amended, is included as the Claims Appendix.

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IV. STATUS OF AMENDMENTS

This application is a divisional of prior application No. 09/574,538, filed May 18, 2000, now U.S. Patent No. 6,331,354. The first Office Action was mailed on March 27, 2003. In response to the first Office Action, appellants mailed a response without claim amendments on June 10, 2003. A second nonfinal Office Action was mailed on September 16, 2003. In response to the second Office Action, appellants mailed an Amendment on December 12, 2003. The Amendment has been entered. The final Office Action was mailed on March 9, 2004. In response to the final Office Action, appellants mailed Amendment B on May 10, 2004, amending Claim 1 to its original language. Amendment B appears to have been entered, as the Advisory Action mailed on June 8, 2004, indicated the rejection of Claims 1-11 under 35 U.S.C. § 112, first paragraph, has been overcome. A Notice of Appeal was filed on July 6, 2004. The Appeal Brief was filed on October 6, 2004. A Notification of Non-Compliant Appeal Brief was mailed on January 11, 2005.

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V. SUMMARY OF CLAIMED SUBJECT MATTER

Claim 1 is the only independent claim. All other claims depend from Claim 1. Claims 3, 7, 8, and 11 are argued separately under Section 112 and 103(a). Claims 7 and 8 are argued separately. The present invention of Claim 1 is related to a process for treating pulp to render it useful for making lyocell fibers.

Pulp is a mass of liberated fibers derived from plant material, especially trees. Depending on the method used in producing the pulp, pulp comprises cellulose, hemicellulose, and lignin in varying proportions. Cellulose is the predominant and most useful component of pulp. Pulp begins as wood that is mechanically refined, chemically digested, or a combination of both mechanical and chemical processing are used to liberate the individual pulp fibers from the wood. Thereafter, the pulp can be chemically treated to remove undesirable coloring components in a process referred to as "bleaching." In the most widely used chemical digestion process, known as the "Kraft" process, wood is treated under alkaline conditions with sodium sulfide. (Page 3, lines 22-30.) In the conventional Kraft process hemicellulose is retained within the pulp.

In the past, cellulose was either derivatized or complexed to render the cellulose capable of being dissolved for use in "spinning," a process for extrusion of a cellulose solution into a regenerating bath that precipitates the cellulose as continuous filaments. (Page 2, lines 21-30.) In recent years, attempts have been made to identify solvents that are capable of dissolving underderivatized cellulose to form dope (a solution of cellulose in water and solvent). A particularly useful class of solvents being used today are the amine N-oxides, particularly the tertiary amine N-oxides. With the advent of amine N-oxide solvents, an alternative now exists to the derivatization or complexing of cellulose.

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"Lyocell" is the accepted generic term for a fiber composed of cellulose precipitated from solution without substitution of hydroxyl groups and formation of chemical intermediates. (Page 3, lines 1-13.) Lyocell fibers are typically produced from wood pulps that have been extensively processed to remove non-cellulose components, especially hemicellulose. It is generally believed that the higher the ratio of non-cellulose components present in dope, the less suitable the dope will be for spinning into lyocell fibers. These highly processed pulps are referred to as dissolving grade, or high alpha pulps, where the term "alpha" refers to the percentage of cellulose. Thus, a high alpha pulp contains a high percentage of cellulose and a correspondingly low percentage of other components, especially hemicellulose. The processing required to generate a high alpha pulp significantly adds to the cost of lyocell fibers and products manufactured from lyocell fibers. (Page 3, lines 14-21.)

To prepare high alpha pulps by the Kraft process, it is necessary to treat the wood with acid before pulping. A significant amount of material, primarily hemicellulose, is solubilized in this acid treatment. Thus, the acid treatment of pulp results in a drop in yield due primarily to the loss of hemicellulose. (Page 4, lines 10-17.)

One measure of the suitability of a pulp useful for lyocell fiber production is its "copper" number. The "copper" number of a pulp is proportional to the carbonyl groups present in the cellulose. A low copper number is a desirable property of pulp that is to be used to make lyocell fibers because it is generally believed that pulps with relatively high copper numbers degrade the amine oxide solvent used to dissolve the cellulose. (Page 4, lines 9-16.)

Transition metals in pulp are also undesirable because transition metals tend to cause degradation of both cellulose and solvent. (Page 4, lines 17-19.)

The present invention of Claim 1 is related to a process that can produce comparatively inexpensive pulps as compared with high alpha pulps. The pulps made according to the process

of the invention have a high hemicellulose content, i.e., are low alpha pulps, but are nevertheless suitable for dissolving and spinning into lyocell fibers. The pulps made by the process of the invention also have acceptable levels of transition metals and a relatively low copper number. Furthermore, the process of the invention can be performed using the pulps made by the conventional Kraft process without the need to perform the acid treatment step. By avoiding an acid treatment step prior to pulping, the overall cost of producing the pulp is reduced. Further, by avoiding the acid treatment, the degradation of hemicellulose is reduced and the overall yield is increased. (Page 15, lines 1-4.)

The process of the invention is performed in a reactor operated at conditions that result in a reduction of the average degree of polymerization of the cellulose in pulp. (Page 18, lines 12-18.) Degree of polymerization (D.P.) is the number of glucose monomers in a cellulose polymer. (Page 15, lines 19 to page 16, line 26.) The process furthermore does not result in any significant decrease in hemicellulose or cause a significant increase in the copper number. The invention of Claim 1 "reduces the average degree of polymerization of cellulose . . . , without substantially reducing the hemicellulose content of the pulp or substantially increasing the copper number." The phrase, "without substantially increasing the copper number" means without increasing the copper number by more than about 100%, preferably not more than about 50%, and most preferably not more than about 25% during the D.P. reduction step. (Page 16, lines 15-26.) Claim 7 places a limit on the increase in copper number at less than 50%. Claim 8 places a limit on the increase in copper number at less than about 25%. The phrase, "without substantially reducing the hemicellulose content" means without reducing the hemicellulose content by more than about 50%, preferably not more than about 15%, and most preferably not more than about 5% during the D.P. reduction step. (Page 15, lines 24-27.)

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The present invention treats a conventional alkaline pulp, such as a Kraft pulp, having a hemicellulose of at least 7%, under alkaline conditions with an amount of an oxidant sufficient to reduce the average degree of polymerization of the cellulose to within the range of from about 200 to about 1100. Lowering the D.P. of the cellulose thereby results in a reduced viscosity of dope that consequently can be spun into lyocell fibers. (Page 16, lines 7-12.) The oxidant used to reduce the D.P. can be any oxidant containing a peroxide group, such as hydrogen peroxide, oxygen, chlorine dioxide, and ozone.

Prior art processes intentionally sought to remove transition metals during bleaching because it was believed that the presence of transition metals resulted in decomposition of hydrogen peroxide into cellulose-degrading intermediates that negatively impacted the viscosity of the cellulose. Unlike the prior art, appellants have discovered that they can take advantage of the presence of naturally occurring transition metals in the wood to partially degrade the hydrogen peroxide to produce intermediates that react with the cellulose to reduce its average degree of polymerization without substantially decreasing the hemicellulose content or increasing the kappa number.

Also, prior art processes have introduced magnesium sulfate as a means of inhibiting the degradation of cellulose. Unlike these processes, appellants prefer not to introduce magnesium sulfate into the reactor or upstream therefrom so that the pulp is contacted with the oxidant in the substantial absence of an inhibitor to the degradation of the cellulose. This advantage is claimed in Claim 11 that recites, "wherein the contacting step occurs in the substantial absence of an inhibitor to the degradation of the cellulose by the oxidant." If magnesium sulfate is present in the pulp prior to the reactor, it is preferred that the ratio of magnesium to the transition metals be less than 50% on a weight percent basis. (Page 20, lines 16-31.) This advantage is claimed in Claim 3 that recites, "wherein the reduction in the average degree of polymerization of the

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cellulose occurs in the presence of a ratio of magnesium to transition metals of less than about 50%."

The absence of an inhibitor and having reduced amounts of magnesium sulfate is an advantage in the claimed process, since the claimed process seeks to degrade the cellulose molecules by reducing the degree of polymerization of the cellulose.

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VI. GROUNDS OF REJECTION TO BE REVIEWED ON APPEAL

- (1) Whether Claims 1-11 are indefinite under 35 U.S.C. § 112.
- (2) Whether Claims 1-11 are unpatentable under 35 U.S.C. § 103(a) over U.S. Patent No. 6,042,769 (Gannon), in view of WO 99/16960 (Stephens) and U.S. Patent No. 5,985,097 (Samuelsson), with U.S. Patent No. 6,210,801 (Luo).
- (3) Whether Claims 1-11 are unpatentable under 35 U.S.C. § 103(a) over U.S. Patent No. 6,042,769 (Gannon), in view of WO 99/16960 (Stephens) and U.S. Patent No. 5,985,097 (Samuelsson), without U.S. Patent No. 6,210,801 (Luo).

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VII. ARGUMENT

A. Rejection of Claims 1-11 Under 35 U.S.C. § 112, Second Paragraph

Claims 1-11 are rejected under 35 U.S.C. § 112, second paragraph, as being indefinite for failing to particularly point out and distinctly claim the subject matter which applicants regard as the invention. Appellants traverse the rejection for the following reasons.

The Examiner has stated at page 4 of the Office Action mailed on March 9, 2004 (hereinafter "Office Action") that "[t]he term 'without substantial increasing of the copper number' when read in view of the specification includes increases up to 100%. A doubling of the copper number is a substantial increase in copper number. Thus the term is indefinite. The argument that a 100% increase is not a substantial increase is not convincing. A 10% increase would be a substantial increase, a 100% increase is an extremely large increase and would not be considered 'without substantial increase.'"

The Court of Appeals for the Federal Circuit has determined that definiteness of claim language must be analyzed, not in a vacuum, but in light of (1) the content of the particular application disclosure, (2) the teachings of the prior art, and (3) the claim interpretation that would be given by one possessing the ordinary level of skill in the pertinent art at the time the invention was made. *See, e.g., In re Marosi, Stabenow, and Schwarzman*, 710 F.2d 799, 803, 218 U.S.P.Q. 289, 291 (Fed. Cir. 1983).

Appellants submit the meaning of the phrase "without substantially increasing the copper number" is not indefinite under 35 U.S.C. § 112, second paragraph. Appellants submit that the phrase "without substantially increasing the copper number" is clear to a person of ordinary skill, either based on the plain and ordinary meaning of the words themselves, and also by referring to the present specification.

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The term "substantially" is often used in conjunction with another term to describe a particular characteristic of the claimed invention. It can be a broad term. *In re Nehrenberg*, 280 F.2d 161, 165, 126 U.S.P.Q. 383, 386 (C.C.P.A. 1960). The limitation "to substantially increase the efficiency of the compound as a copper extractant" was definite in view of the general guidelines contained in the specification. *In re Mattison*, 509 F.2d 563, 565, 184 U.S.P.Q. 484, 486-87 (C.C.P.A. 1975). The court in *Andrew Corp. v. Gabriel Electronics* held that the limitation "which produces substantially equal E and H plane illumination patterns" was definite because one of ordinary skill in the art would know what was meant by "substantially equal." *Andrew Corp. v. Gabriel Electronics*, 847 F.2d 819, 823-824, 6 U.S.P.Q.2d 2010, 2013-14 (Fed. Cir. 1988).

Words of degree, such as "substantially," are addressed in the case, *Exxon v. United States*, 265 F.3d 1371, 60 U.S.P.Q.2d 1272 (Fed. Cir. 2001). In this Federal Circuit case, the phrase "for a period sufficient to increase substantially the initial catalyst activity" did not render the claim invalid for indefiniteness, because a person of ordinary skill would understand what the patentee intended, even though the patent did not specify which method was used to calculate the increase in productivity. *Id.* at 1375, 1278.

As the above cases distinctly point out, the term "substantially" is not indefinite if one of ordinary skill in the art would understand what was meant by the phrase at issue. It is apparent that the Examiner has understood the phrase by referring to the specification. It is, therefore, apparent that a person of ordinary skill would also understand the phrase by referring to the specification.

The Examiner is imposing a subjective opinion that the phrase is indefinite because the meaning is supposedly contrary to an "accepted" meaning. See p. 4 of the Office Action mailed on September 16, 2003, where the Examiner states "[t]he argument that Applicant is allowed to define terms is correct. However, the definitions must be within that conventionally used in the

art. The art would not recognize a doubling of copper number to be "without substantial increasing." However, there is no evidence in the record other than the Examiner's unsupported assertion of what is the commonly accepted meaning of "substantially." Appellants submit that there can be no prior commonly accepted meaning of the phrase because the inventive process is novel, and the phrase is one of degree. Regardless, appellants have unambiguously defined, "without substantially increasing the copper number," in the specification to be without increasing the copper number by more than about 100%, preferably not more than about 50% and most preferably not more than about 25% during the D.P. reduction step. Please see the present specification on page 16, lines 8-11. One of ordinary skill would readily understand what is meant by the phrase "without substantially increasing the copper number" by merely referring to the specification. "If the claims, read in light of the specification, reasonably apprise those skilled in the art both of the utilization and scope of the invention, and if the language is as precise as the subject matter permits, the courts can demand no more." *See Hybritech Inc. v. Monoclonal Antibodies, Inc.*, 802 F.2d 1367, 1385, 231 U.S.P.Q. 81, 94-95 (Fed. Cir. 1986), *citing Shatterproof Glass Corp. v. Libbey Owens Ford Co.*, 758 F.2d 613, 624, 225 U.S.P.Q. 634, 641 (Fed. Cir. 1985).

Accordingly, for all the foregoing reasons reversal of the Examiner's rejection of Claims 1-11 under 35 U.S.C. § 112, second paragraph, is respectfully requested.

A.(i). Rejection of Claim 7 Under 35 U.S.C. § 112, Second Paragraph

Claim 7 is being argued separately. Claim 7 is believed to be patentable for the reasons discussed above in Section A. In addition, Claim 7 places a numerical limit of the amount of increase in copper number. Appellants submit that Claim 7 is thus clear and unambiguous on the face of the claim, even without resort to the specification.

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In reviewing a claim for compliance with 35 U.S.C. § 112, second paragraph, the Examiner must consider the claim as a whole to determine whether the claim apprises one of ordinary skill in the art of its scope and, therefore, serves the notice function required by 35 U.S.C. § 112, second paragraph. *See Solomon v. Kimberly-Clark Corp.*, 216 F.3d 1372, 1379, 55 U.S.P.Q.2d 1279, 1283 (Fed. Cir. 2000). Appellants submit that nothing can be more clear and precise when the scope of the term at issue in Claim 7 is apparent on the face of the claim.

A.(ii). Rejection of Claim 8 Under 35 U.S.C. § 112, Second Paragraph.

Claim 8 is being argued separately. Claim 8 is believed to be patentable for the reasons discussed above in Section A. In addition, Claim 8 places a numerical limit of the amount of increase in copper number. Appellants submit that Claim 8 is thus clear and unambiguous on the face of the claim, even without resort to the specification.

In reviewing a claim for compliance with 35 U.S.C. § 112, second paragraph, the Examiner must consider the claim as a whole to determine whether the claim apprises one of ordinary skill in the art of its scope and, therefore, serves the notice function required by 35 U.S.C. § 112, second paragraph. *See Solomon v. Kimberly-Clark Corp.*, 216 F.3d 1372, 1379, 55 U.S.P.Q.2d 1279, 1283 (Fed. Cir. 2000). Appellants submit that nothing can be more clear and precise when the scope of the term at issue in Claim 8 is apparent on the face of the claim.

B. Rejection of Claims 1-11 Under 35 U.S.C. § 103(a) in View of Gannon, Stephens, Samuelsson, and Luo.

Claims 1-11 are rejected under 35 U.S.C. § 103(a) as being unpatentable over Gannon et al. (U.S. Patent No. 6,042,769) in view of Stephens (WO 99/16960) and Samuelsson (U.S. Patent No. 5,985,097) with Luo et al. (U.S. Patent No. 6,210,801). Appellants respectfully traverse the rejection for the following reasons.

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1. Brief overview of the references.

The Gannon reference relates to a process for manufacturing a lyocell fiber having an increased tendency to fibrillate. Fibrillated fibers are advantageous for uses such as hydroentangled fabrics. Gannon describes the conventional lyocell fiber forming process that involves dissolving cellulose with amine N-oxide to form a solution, extruding the solution through a die to form a plurality of filaments, and washing the filaments to remove the solvent, thereby forming lyocell fiber. The characterizing step is described as subjecting the lyocell fiber to conditions effective to reduce the degree of polymerization of the cellulose by at least about 200 units. Col. 2, lines 2-4. Gannon describes reducing the degree of polymerization of cellulose in a lyocell fiber before or after drying, which in all cases is after the formation of the lyocell fiber. Col. 2, lines 12-22. The desired reduction in cellulose degree of polymerization may be carried out by a bleaching treatment. The bleaching treatment liquor may be applied to the fiber by passage through a bath, by padding, or by spraying. The Gannon reference does not, however, describe how to render a pulp that is high in hemicellulose into a pulp that is useful for spinning into lyocell fiber, without substantially increasing its copper number or decreasing its hemicellulose content.

The Stephens reference describes a process whereby the degree of polymerization of a pulp is reduced with a cellulolytic enzyme. The Stephens reference describes converting regular Kraft wood pulp, without having undergone a prehydrolysis step, into a pulp suitable for the manufacture of cellulose derivatives and lyocell fibers. The process relies on the activity of enzymes to reduce the degree of polymerization. The process, however, removes hemicellulose.

The Samuelsson reference is related to a method that suppresses the effects of transition and alkaline earth metal compounds during oxygen bleaching of chemically digested pulps. According to Samuelsson, operating conditions of the oxygen bleaching stage are modified so

that depolymerization of the cellulose is low in this stage. To accomplish this objective, the oxidation states of transition metals are changed. The change in oxidation state can be brought about by changes in the oxygen pressure, alkali concentration and temperature.

The Luo reference is related to a process for reducing the degree of polymerization of cellulose of a pulp using a variety of chemicals. The pulps produced thereby are useful for making lyocell fibers. One of the advantages of lyocell fibers produced by the pulps made according to the Luo reference is the decreased tendency of the lyocell fibers to fibrillate. According to the Luo process, after reducing the degree of polymerization of cellulose, the copper number of the pulp has to be reduced with sodium borohydride or sodium hydroxide.

The Examiner suggests the claimed invention is obvious by combining the Gannon, Stephens, Samuelsson, and Luo references.

An obviousness rejection requires that there be a suggestion or motivation either in the references or in the knowledge that is generally available to modify a reference or to combine references. In addition, there must be a reasonable expectation of success and all the elements of the claims must be taught or suggested by the prior art.

Appellants will show that the Examiner has failed to provide all the elements of a *prima facie* case of obviousness.

2. There is no suggestion or motivation to combine the Stephens reference with the Gannon reference. There is no reasonable expectation of success. The references do not describe all the elements of the claimed invention.

From the brief overview of the Gannon reference above, it is evident that the Examiner has mischaracterized the Gannon reference by suggesting pulp and lyocell fiber mean the same thing, as is evident from the Examiner's statement on page 3 of the Office Action:

Gannon et al. fairly disclose a process for making lyocell fibers comprising the steps of: (a) contacting an alkaline pulp comprising cellulose and hemicellulose under alkaline condition with an amount of oxidant

(hydrogen peroxide or ozone) sufficient to reduce the average degree of polymerization of the cellulose to the range of from about 200 to about 1100 and (b) forming the fibers from the pulp treated in accordance with step (a) (see Cols. 2-6). Gannon et al. teaches the claimed invention except for the limitation of without substantially reducing the hemicellulose content of the pulp or substantially increasing the copper number.

There is no indication by the Examiner where the Gannon reference specifically describes steps (a) and (b), other than to make broad reference to Columns 2 through 6.

Claim 1 recites "contacting an alkaline pulp comprising cellulose and at least about 7% hemicellulose under alkaline conditions with an amount of an oxidant sufficient to reduce the average degree of polymerization of the cellulose to within the range of from about 200 to about 1100, without substantially reducing the hemicellulose content of the pulp or substantially increasing the copper number."

In direct contrast to Claim 1, Gannon describes a lyocell fiber treatment that reduces the lyocell fiber's degree of polymerization by about 200 units. See the Gannon reference's Abstract. A lyocell fiber is described in the SUMMARY OF CLAIMED SUBJECT MATTER section of this appeal brief as being precipitated (i.e., regenerated) from a solution of cellulose, water and a solvent. In direct contrast, pulp that is the object of the present inventive process is the precursor material of lyocell fibers that has not undergone dissolution and consequently is not regenerated. It is the formed lyocell fibers that are described as being treated by the Gannon reference, not the pulp as claimed. (Col. 2, lines 12-22.)

Appellants believe the Examiner's reading of "pulp" in the claims being synonymous with the "lyocell fiber" of Gannon, is in error. Not only do the words themselves define different materials. It can be shown that there are structural differences between the cellulose in pulp and the cellulose in a lyocell fiber. Consequently, a "pulp" and a "lyocell fiber" are recognized in the art as being materially different. In support of appellants' position that pulp and lyocell fiber are

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structurally different, appellants direct the attention of the Board to two excerpts of publications included in the evidence appendix of this appeal brief. The excerpts were included with a response, mailed on May 10, 2004.

At page 53 of the Sjöström publication, the differences of Cellulose I, which is native cellulose, are contrasted with regenerated cellulose, Cellulose II. Furthermore, on page 55, the publication states, "Cellulose II is formed whenever the lattice of Cellulose I is destroyed, for example, on swelling with strong alkali or on dissolution of cellulose." Appellants submit that the Gannon reference describes the treatment of Cellulose II because the Gannon reference explicitly describes the dissolution of cellulose in a solvent followed by regeneration and treatment (Col. 1, line 65). See also the Rydholm publication, at pages 116-17, describing the differences in cellulose between native cellulose (Cellulose I) and regenerated cellulose (Cellulose II). Again, lyocell fiber is a regenerated cellulose.

Accordingly, based on the discussion above, it is not correct to state that Gannon et al. disclose contacting an alkaline pulp under alkaline conditions with an amount of oxidant, when, in fact, what is described by Gannon is treatment of a lyocell fiber. Pulp and lyocell fiber do not mean the same thing.

Furthermore, the Gannon reference is silent on the amount of hemicellulose present in the lyocell fiber and therefore cannot teach the reduction of the D.P. of cellulose without substantially decreasing the hemicellulose content of a pulp with at least 7% hemicellulose. This alone distinguishes the claimed invention over the Gannon reference.

Perhaps appreciating that the Gannon reference does not describe treating a pulp, the Examiner further states that, "[t]he argument that Gannon uses a different starting material than the instant process is not convincing as it would have been obvious to use non-regenerated cellulose from the teachings of WO 99/16960 [Stephens]," and "[i]t would have been *prima facie*

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obvious from the teachings of WO 99/16960 [Stephens] to use alkaline Kraft pulp as the fiber of Gannon et al. The Kraft pulp of WO 99/16960 [Stephens] is the same non-regenerated cellulose used by Applicant."

Before the references can be combined or modified there has to be a suggestion or motivation to do so. "Obviousness cannot be established by combining the teachings of the prior art to produce the claimed invention absent some teaching, suggestion or incentive supporting the combination." *Ecolochem, Inc. v. Southern California Edison*, 227 F.3d 1361; 1372, 56 U.S.P.Q.2d 1065, 1073 (Fed. Cir. 2000), *citing ACS Hosp. Sys., Inc. v. Montefiore Hosp.*, 732 F.2d 1572, 1577, 221 U.S.P.Q. 929, 933 (Fed. Cir. 1984). If the motivation is not immediately apparent, it is the duty of the Examiner to explain why the combination of the teachings is proper. (*Ex parte Skinner*, 2 U.S.P.Q.2d 1788, 1790 (Bd. Pat. App. & Inter. 1986).

Broad allegations of obviousness and of the teachings of Stephens reference do not further prosecution. No explanation is provided why the modification of Gannon with Stephens would be obvious. Furthermore, any attempt at combining and/or modifying the Gannon reference with the Stephens reference makes no sense. The Gannon reference is directed at treating lyocell fibers. The Stephens reference is directed at treating pulp.

Is the Examiner implying that prior to forming the lyocell fibers, Gannon should treat the pulp? Or is the Examiner implying that Stephens could substitute bleach liquor for enzymes? Either way, modification of the Gannon or Stephens reference involves a complete change in the principle of the operation of the Gannon and Stephens inventions. The Gannon reference's objective is to produce a lyocell fiber with an increased tendency to fibrillate. (See Col. 1, lines 61-64). Appellants submit the Gannon reference teaches away from reducing the D.P. of pulp. Gannon mentions that the cellulose concentration in dope prior to spinning is directly proportional to the tendency of lyocell fibers to fibrillate. However, increasing the cellulose

concentration requires reducing the D.P. of cellulose to maintain the dope viscosity below the maximum working viscosity. Gannon does not see this as a viable alternative to his method. The increase in fibrillation tendency achievable by use of the Gannon process is generally greater than the increase achievable by raising the cellulose concentration of the solution. (See Col. 5, lines 1-4).

The combination of Gannon with Stephens does not result in the claimed invention. If one were to combine Stephens with Gannon, the pulp used by Gannon would be substituted with the pulp used by Stephens. The pulp would then be dissolved in a solvent, and extruded. However, as taught by Sjöström, the dissolution process would result in cellulose II fiber before it was treated by the process of Gannon. Once again, as stated at the very beginning of this section, lyocell fiber and pulp are not the same. Therefore, combination of Gannon with Stephens does not result in the claimed invention.

As discussed above, the Gannon reference is specifically directed at treating the formed lyocell fibers, not the pulp. Combining or modifying the Gannon reference with the Stephens reference would not only change the principle of operation of the Gannon reference, but also would appear to render the Gannon reference unsatisfactory for its intended purpose. If the Stephens method is modified by incorporating the Gannon method, that is a complete revision of the Stephens invention. If the proposed modification renders the prior art invention being modified unsatisfactory for its intended purpose, there is no suggestion or motivation to make the proposed modification. *In re Gordon*, 733 F.2d 900, 902, 221 U.S.P.Q. 1125, 1127 (Fed. Cir. 1125). If the proposed modification or combination of the prior art would change the principle of operation of the prior art invention being modified, then the teachings of the references are not sufficient to render the claims obvious. *In re Ratti*, 270 F.2d 810, 813, 123 U.S.P.Q. 349, 352 (C.C.P.A. 1959).

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Finally, the invention defined by Claim 1 recites that the pulp that undergoes the D.P. reduction process has at least a 7% hemicellulose content that is not substantially reduced at the time of reducing the D.P. of the cellulose. The combination of the Gannon and the Stephens references fails to teach a process for reducing the D.P. of cellulose that also does not result in any substantial reduction in hemicellulose content of a pulp having at least 7% hemicellulose or any substantial increase in copper number. Contrary to the invention defined by Claim 1, the Stephens reference describes the removal of hemicellulose, not an attempt at avoiding its reduction. See page 4, lines 15-17; page 8, lines 9-11; and page 8, lines 19-21 in the Stephens reference.

Because at the very least, there is no suggestion or motivation to combine the Stephens reference with the Gannon reference, no reasonable expectation of success, and a clear lack of all of the elements of Claim 1, the Examiner has not established a *prima facie* case of obviousness. The deficiencies in the Gannon and Stephens references are not cured by either Samuelsson, Luo, or both as further discussed below. Accordingly, the reversal of the Examiner's rejection of Claim 1 and dependent Claims 2-11 is respectfully requested.

3. There is no suggestion or motivation to combine the Samuelsson reference with the Gannon reference. There is no reasonable expectation of success. The references do not describe all the elements of the claimed invention.

At page 3 of the Office Action, the Examiner states:

Samuelsson et al. teaches that the catalysis of the depolymerization of cellulose and hemicellulose during peroxide bleaching can be controlled by monitoring and controlling the ratio of transition metals and Mg in the pulp (Col. 4, lines 10-22 and 45-47, of the Samuelsson reference). It would have been obvious to prevent the degradation of the hemicellulose and cellulose in the cellulosic material of Gannon et al. by adding the proper amount of Mg as taught by Samuelsson et al. Obviously such addition would avoid substantial reduction of the hemicellulose content, which would not be degraded.

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The Samuelsson reference is directed to a modification of an oxygen bleaching process to prevent the depolymerization of cellulose with the addition of magnesium and manganese. See Col. 2, lines 37-47. The only reference to hemicellulose appears in Col. 1, line 35. This brief mention of hemicellulose only addresses the issue that metals in pulp leads to a depolymerization of hemicellulose. The Samuelsson reference never teaches whether degradation of hemicellulose could or should be prevented. The Samuelsson reference's main focus is directed towards preventing the depolymerization of cellulose. See Col. 3, lines 24-27.

The Samuelsson reference's objective of preventing the depolymerization of cellulose is directly contrary to the objective of Gannon which is to reduce the degree of polymerization of cellulose to increase the tendency of a lyocell fiber to fibrillate. There is no reason why Gannon should prevent depolymerization of cellulose when the explicit teaching of Gannon is to cause depolymerization of the cellulose. (See Col. 2, lines 3-5.) Accordingly, there is nothing to suggest or motivate one to combine or modify Gannon in a way based on Samuelsson.

Also, in direct contrast to the Samuelsson reference, Claim 1 specifically recites reducing the degree of polymerization of cellulose . . . without substantially reducing the hemicellulose content or increasing the copper number. Not only is there no suggestion or motivation to combine the Samuelsson reference with the Gannon reference, but any attempt to do so is bound to fail because implementing the suggestion to prevent the degradation of hemicellulose by adding magnesium will necessarily also lead to preventing the degradation of cellulose. The prevention of cellulose degradation is both contrary to the Gannon reference and Claim 1. In the SUMMARY OF THE CLAIMED SUBJECT MATTER, appellants have pointed out that the prior art teaches the introduction of magnesium sulfate as a means to inhibit the degradation of cellulose, whereas appellants find it advantageous not to introduce magnesium sulfate, and further describe that if magnesium sulfate is naturally present in the pulp, its content is limited to

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less than a specific ratio. The object of the Gannon reference is to decrease the degree of polymerization of cellulose, not suppress cellulose depolymerization which is what will occur if magnesium compounds are added, as taught by Samuelsson. Consequently, adding magnesium to the method described by Gannon leaves this prior art invention inoperative. When the prior art teaches away from the claimed invention, the invention is not obvious in view thereof. *In re Sponnoble*, 405 F.2d 578, 588, 160 U.S.P.Q. 237, 244 (C.C.P.A. 1969) ("a combination of [references] would produce a seemingly inoperative device." Parenthetical added).

There is no suggestion or motivation to combine or modify Gannon based on Stephens. There is no reasonable expectation of success by combining Gannon with Stephens. The combination of Gannon with Stephens fails to provide all elements of the claimed invention. The suggestion or motivation to combine or modify Gannon with Stephens, and Samuelsson is not found in Samuelsson. Samuelsson teaches away from combining with Gannon. The combination of Samuelsson with Gannon and Stephens does not yield a reasonable expectation of success nor does the combination of Gannon, Stephens, and Samuelsson provide all the elements of the claimed invention.

4. There is no suggestion or motivation to combine the Luo reference with the Gannon reference. There is no reasonable expectation of success. The references do not describe all the elements of the claimed invention.

At pages 3-4 of the Office Action, the Examiner states that,

[s]ince the copper number is directly related to the cellulose degradation, see specification, page 16, lines 8-20, it would have been obvious that preventing cellulose degradation by adding Mg to the pulp during peroxide bleaching as taught by Gannon et al., would prevent an increase in the copper number.

Before appellants address the errors with combining the Gannon and Luo references, appellants wish to correct what appears to be a misunderstanding on the part of the Examiner. The passage taken from appellants' specification (page 16, lines 8-20) is in the context of

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dissolving the cellulose in the amine oxide solvent. That passage explains that pulp having a high copper number may cause undesirable degradation of the cellulose and solvent during and after dissolution of the pulp to form a dope. The passage does not relate to the step in appellant's claims of reducing the degree of polymerization without substantially increasing the copper number. To correct for the undesirable feature of pulps with high copper numbers, appellants' method yields a pulp with a low copper number.

There is no teaching from the references so far discussed above to reduce the degree of polymerization of cellulose without substantially increasing the copper number at the time of reducing the degree of polymerization.

The Examiner states at page 4 of the Office Action that:

[i]f necessary, it would have been obvious to prevent an increase in the copper number by treating the lyocell with sodium borohydride to decrease the copper number as taught by Luo et al.

Again, a broad allegation that something is obvious is meaningless without a suggestion or motivation to combine references. There is no suggestion or motivation to combine the Gannon reference with the Luo reference.

The Luo reference describes the reduction of the D.P. of cellulose of pulp, followed by the reduction in copper number. As explained in THE SUMMARY OF THE CLAIMED INVENTION, a low copper number is a desirable characteristic in a pulp that is to be dissolved, because a pulp with a relatively high copper number degrades the solvent. Appellants fail to see the value of reducing the copper number of Gannon's lyocell fibers when the fibers have already been dissolved, spun and formed.

Even if the Luo and Gannon references are combined, the resultant process is not the claimed process. Claim 1, paraphrased in part, requires prevention of a substantial increase in copper number during the treatment to reduce the degree of polymerization. See the

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SUMMARY OF THE CLAIMED INVENTION. Luo describes a two-step process. The first step reduces the D.P. of the cellulose of the pulp. The second step reduces the copper number of the cellulose of the pulp. The second step is necessary in the Luo reference because the reduction in the D.P. of cellulose causes an increase in copper number. See Col. 13, lines 31-39, of the Luo reference. Appellants' claimed invention eliminates the need to have a separate, distinct step for reducing the copper number. Accordingly, the Luo et al. reference does not describe a step to "reduce the average degree of polymerization . . . without . . . substantially increasing the copper number," as recited by Claim 1. Elimination of an element (or step) with retention of its function is an indicia of unobviousness. *In re Edge*, 359 F.2d 896, 899, 149 U.S.P.Q. 556, 557 (C.C.P.A. 1966).

Furthermore, one of the advantages described by Luo, is the decreased tendency of lyocell fibers to fibrillate. See the Abstract. The Gannon reference's objective is to increase the fibrillation tendency of lyocell fibers. Accordingly, there is no suggestion or motivation to combine the Luo reference with the Gannon reference.

There is no suggestion or motivation to combine or modify Gannon based on Stephens and Samuelsson. There is no reasonable expectation of success by combining Gannon with Stephens and Samuelsson. The combination of Gannon with Stephens and Samuelsson fails to provide all the elements of the claimed invention. The suggestion or motivation to combine or modify Gannon with Stephens, Samuelsson, and Luo is not found in Luo. Luo teaches away from combining with Gannon. The combination of Luo with Gannon, Stephens, and Samuelsson does not yield a reasonable expectation of success nor does the combination of Gannon, Stephens, Samuelsson, and Luo provide all the elements of the claimed invention.

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B.(i). The Rejection of Claim 3 Under 35 U.S.C. § 103(a) in View of Gannon, Stephens, Samuelsson, and Luo.

Claim 3 is being argued separately. Claim 3 is believed patentable for the reasons discussed above in Section B. In addition, Claim 3 recites, "wherein the reduction in the average degree of polymerization of the cellulose occurs in the presence of a ratio of magnesium to transition metals of less than about 50%".

As explained in the SUMMARY OF THE CLAIMED INVENTION, appellants have found it advantageous not to add magnesium sulfate or other inhibitor that prevents the depolymerization of cellulose. The reason for not having an inhibitor or having a limit on the amount of magnesium is because the claimed invention seeks to advantageously reduce the D.P. of cellulose of a pulp to within a range that can produce a dope that can be spun into lyocell fiber. Any inhibitor, or magnesium compound would not allow the reduction of the D.P. of cellulose. There is no teaching or suggestion in the references discussed above to perform a method in accordance with Claim 3. In direct contrast, the only reference that appears to address the issue of magnesium is the Samuelsson reference. However, the Samuelsson reference describes the addition of magnesium compounds (inhibitors) to prevent depolymerization. Appellants reduce or eliminate the amount of magnesium and inhibitor to achieve the opposite result, a reduction in the D.P. of cellulose. Accordingly, Claim 3 is patentable in view of Gannon, Stephens, Samuelsson, and Luo, taken alone or in combination.

B.(ii). The Rejection of Claim 11 Under 35 U.S.C. § 103(a) in View of Gannon, Stephens, Samuelsson, and Luo.

Claim 11 is being argued separately. Claim 11 is believed patentable for the reasons discussed above in Section B. In addition, Claim 11 recites, "wherein the contacting step occurs in the substantial absence of an inhibitor to the degradation of the cellulose by the oxidant."

As explained in the SUMMARY OF THE CLAIMED INVENTION, appellants have found it advantageous not to add magnesium sulfate or other inhibitor that prevents the

depolymerization of cellulose. The reason for not having an inhibitor or having a limit on the amount of magnesium is because the claimed invention seeks to advantageously reduce the D.P. of cellulose of a pulp to within a range that can produce a dope that can be spun into lyocell fiber. Any inhibitor, or magnesium compound would not allow the reduction of the D.P. of cellulose. There is no teaching or suggestion in the references discussed above to perform a method in accordance with Claim 11. In direct contrast, the only reference that appears to address the issue of magnesium is the Samuelsson reference. However, the Samuelsson reference describes the addition of magnesium compounds (inhibitors) to prevent depolymerization. Appellants reduce or eliminate the amount of magnesium and inhibitor to achieve the opposite result, a reduction in the D.P. of cellulose. Accordingly, Claim 11 is patentable in view of Gannon, Stephens, Samuelsson, and Luo, taken alone or in combination.

B.(iii) The Rejection of Claim 7 Under 35 U.S.C. § 103(a) in View of Gannon, Stephens, Samuelsson, and Luo.

Claim 7 is being argued separately. Claim 7 is believed patentable for the reasons discussed above in Section B. In addition, Claim 7 recites "wherein the copper number increases less than 50%."

Of all the references that were cited and applied, the only reference that addresses copper number is the Luo reference. However, the Luo reference describes that the increase in copper number cannot be prevented during the D.P. reduction step, and finds it necessary to perform a second and subsequent step to reduce the copper number. In contrast, the invention defined by Claim 7, advantageously holds down the copper number increase to a small percentage that avoids the need for a second subsequent step to reduce the copper number. Accordingly, Claim 7 is patentable in view of Gannon, Stephens, Samuelsson, and Luo, taken alone or in combination.

B.(iv) The Rejection of Claim 8 Under 35 U.S.C. § 103(a) in View of Gannon, Stephens, Samuelsson, and Luo.

Claim 8 is being argued separately. Claim 8 is believed patentable for the reasons discussed above in Section B. In addition, Claim 8 recites, "wherein the copper number increases less than about 25%."

Of all the references that were cited and applied, the only reference that addresses copper number is the Luo reference. However, the Luo reference describes that the increase in copper number cannot be prevented during the D.P. reduction step, and finds it necessary to perform a second and subsequent step to reduce the copper number. In contrast, the invention defined by Claim 8 advantageously holds down the copper number increase to a small percentage that avoids the need for a second subsequent step to reduce the copper number. Accordingly, Claim 8 is patentable in view of Gannon, Stephens, Samuelsson, and Luo, taken alone or in combination.

C. The Rejection of Claims 1-11 Under 35 U.S.C. § 103(a) in View of Gannon, Stephens, and Samuelsson

Claims 1-11 are rejected under 35 U.S.C. § 103(a) as being unpatentable over Gannon et al. (U.S. Patent No. 6,042,769) in view of Stephens (WO 99/16960) and Samuelsson (U.S. Patent No. 5,985,097) without Luo et al. (U.S. Patent No. 6,210,801). Appellants respectfully traverse the rejection for the following reasons.

1. There is no suggestion or motivation to combine Gannon, Stephens, and Samuelsson. There is no reasonable expectation of success. The references do not describe all the elements of the claimed invention.

An obviousness rejection requires that there be a suggestion or motivation either in the references or in the knowledge that is generally available to modify a reference or to combine references. In addition, there must be a reasonable expectation of success, and all the elements of the claims must be taught or suggested by the prior art.

As discussed above in sections B.1. through B.3., there is no suggestion or motivation to combine or modify Gannon with Stephens and Samuelsson to yield appellants' invention of Claims 1-11. Appellants incorporate the previous argument of Section B into this section. In addition, appellants submit that Gannon, Stephens, and Samuelsson fail to teach or suggest, "to reduce the average degree of polymerization of the cellulose . . . without . . . substantially increasing the copper number.

The Examiner does not rely on the teachings of Luo for this ground of rejection. Rather, the Examiner relies on appellants' own specification to suggest that, "[i]t would have been obvious that preventing cellulose degradation by adding Mg to the pulp during peroxide bleaching as taught by Gannon et al., would prevent an increase in the copper number."

At pages 3-4 of the Office Action, the Examiner states that,

[s]ince the copper number is directly related to the cellulose degradation, see specification, page 16, lines 8-20, it would have been obvious that preventing cellulose degradation by adding Mg to the pulp during peroxide bleaching as taught by Gannon et al., would prevent an increase in the copper number.

The Examiner's interpretation of appellants' own disclosure is in error.

Furthermore, appellants point out that Claim 1 recites "to reduce the average degree of polymerization of the cellulose," which is in direct contrast to preventing cellulose degradation by the addition of Mg. The passage at page 16, lines 8-20 of appellants' specification explains that pulp having a high copper number may cause degradation of the cellulose and solvent during and after dissolution of the pulp to form a dope. The passage does not relate to the step in appellants' claims of reducing the degree of polymerization without substantially increasing the copper number. The relevant sentences in the passage state "[t]he degree to which the copper number changes during the D.P. reduction step is determined by comparing the copper number of the pulp entering the D.P. reduction step and the copper number of the treated pulp after the D.P.

reduction step. A low copper number is desirable because it is generally believed that a high copper number causes cellulose and solvent degradation during and after dissolution of the treated pulp to form dope." Therefore, the reduction of the average degree of polymerization of the cellulose without an increase in copper number recited in Claim 1 is not related to the degradation of cellulose and solvent discussed at page 16, lines 8-20.

In the previous ground for rejection, appellants argued against combining Luo with Gannon for lack of suggestion or motivation to do so, and even if combined, the references failed to teach or suggest all limitations. As is evident from appellants' explanation of the passage in appellants own application, the Examiner has misconstrued appellants' specification. Accordingly, there is no suggestion or motivation "to reduce the degree of polymerization . . . without . . . substantially increasing the copper number."

Accordingly, because of the error in applying appellants' own application to reject Claims 1-11, appellants respectfully request the reversal of this ground for rejection of Claims 1-11.

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VIII. CONCLUSION

In view of the above remarks, appellants respectfully submit that each of Claims 1-11 is patentable over the references of record. A decision reversing the Examiner's rejections and finding all pending claims to be in condition for allowance is respectfully requested.

Respectfully submitted,

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I hereby certify that this correspondence is being deposited in triplicate with the U.S. Postal Service in a sealed envelope as first class mail with postage thereon fully prepaid and addressed to Mail Stop Appeal Brief - Patents, Commissioner for Patents, P.O. Box 1450, Alexandria, VA 22313-1450, on the below date.

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CLAIMS APPENDIX

1. (Previously presented) A process for making a composition for conversion to lyocell fiber, said process comprising:

contacting an alkaline pulp comprising cellulose and at least about 7% hemicellulose under alkaline conditions with an amount of an oxidant sufficient to reduce the average degree of polymerization of the cellulose to within the range of from about 200 to about 1100, without substantially reducing the hemicellulose content of the pulp or substantially increasing the copper number.

2. (Original) The process of Claim 1 wherein said oxidant comprises at least one member of the group consisting of a chemical with a peroxide group, oxygen, chlorine dioxide, ozone and combinations thereof.

3. (Original) The process of Claim 2 wherein the reduction in the average degree of polymerization of the cellulose occurs in the presence of a ratio of magnesium to transition metals of less than about 50%.

4. (Original) The process of Claim 1 wherein the hemicellulose content of the pulp is reduced less than about 50%.

5. (Original) The process of Claim 4 wherein the hemicellulose content of the pulp is reduced less than about 15%.

6. (Original) The process of Claim 4 wherein the hemicellulose content of the pulp is reduced less than about 5%.

7. (Original) The process of Claim 1 wherein the copper number increases less than 50%.

8. (Original) The process of Claim 1 wherein the copper number increases less than about 25%.

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9. (Original) The process of Claim 2 wherein the contacting step further comprises contacting the pulp with an alkali source selected from the group consisting of sodium hydroxide, oxidized white liquor, and unoxidized white liquor.

10. (Original) The process of Claim 1, wherein the alkaline pulp and oxidant are contacted at a pH greater than about 8.0.

11. (Original) The process of Claim 1, wherein the contacting step occurs in the substantial absence of an inhibitor to the degradation of the cellulose by the oxidant.

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EVIDENCE APPENDIX

A. Rydholm, Sven A., *Pulping Processes*, InterScience Publishers, 1965, pp. 112-119.

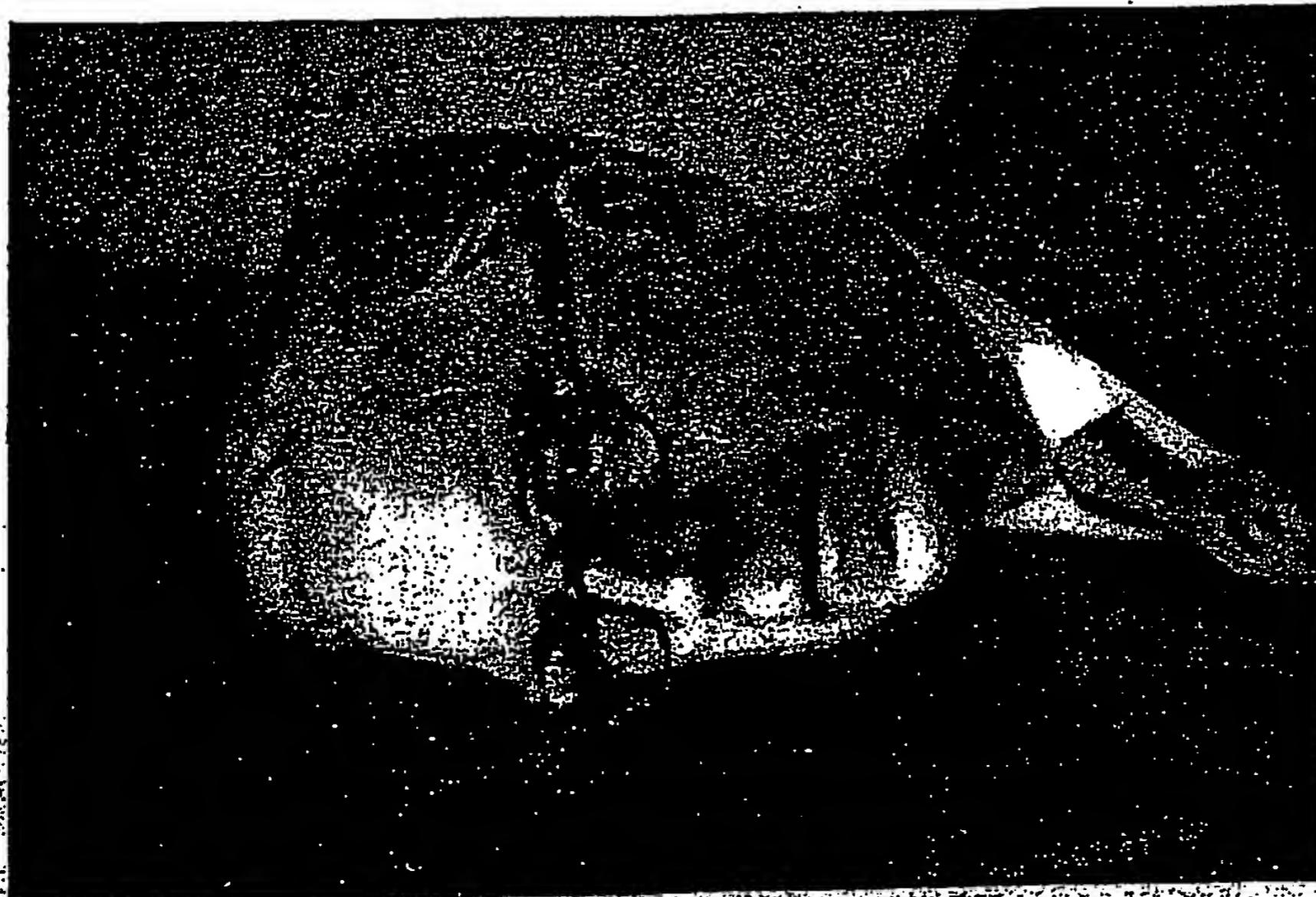
B. Sjöström, Eero, *Wood Chemistry: Fundamentals and Applications*, Academic Press, 1981, pp. 52-57.

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Pulping Processes

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Pioneer of modern pulping chemistry

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same width as the elementary fibrils (270, 052, 049), figure 4.0 (270). The average length of these fragments depends on the pretreatment, those pulps which have been alkali-treated yielding shorter micelles on hydrolysis than those which have not (414). In the former case, the micelles are about 500–1,000 Å long, corresponding to the length of molecules of DP 100–200, which is also the average DP of the hydrolyzed pulp (70). Thus the hydrolysis of cellulose suspensions is pronouncedly heterogeneous, and it is assumed that the amorphous regions form the starting points of this

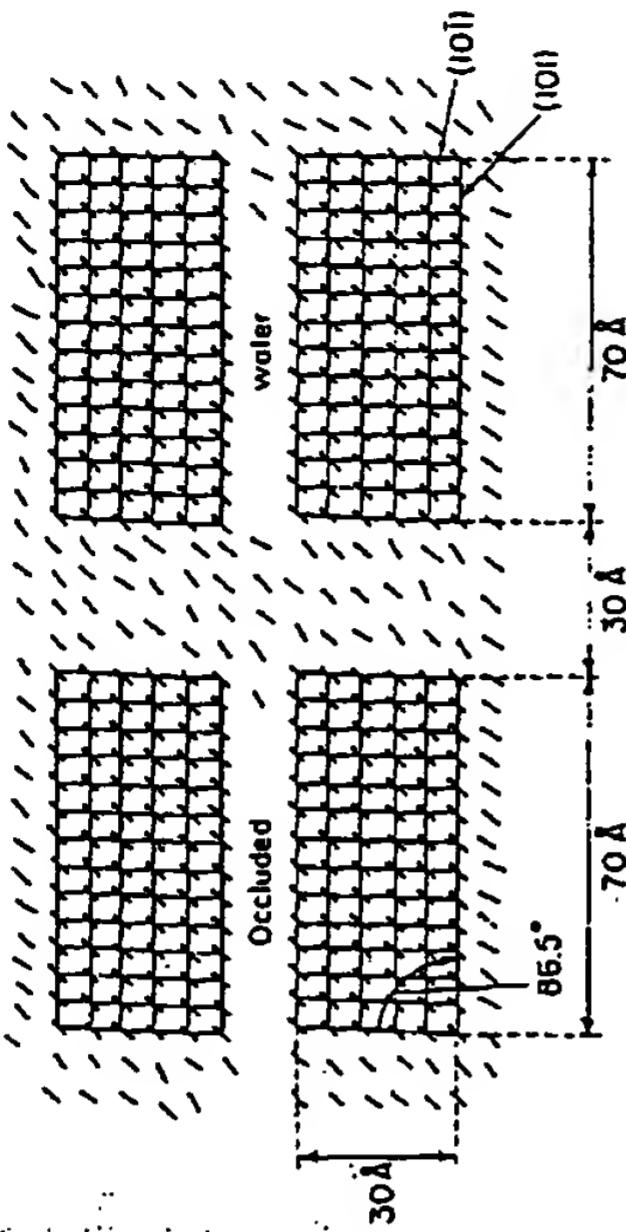


Fig. 4.7. Diagrammatic representation of a cross-section of a microfibril as an aggregation of elementary fibrils (Frey-Wyssling)



Fig. 4.8. Micelles from hydrolysed ramie cellulose, deposited on a glass surface (x 47,000) (Morthead)

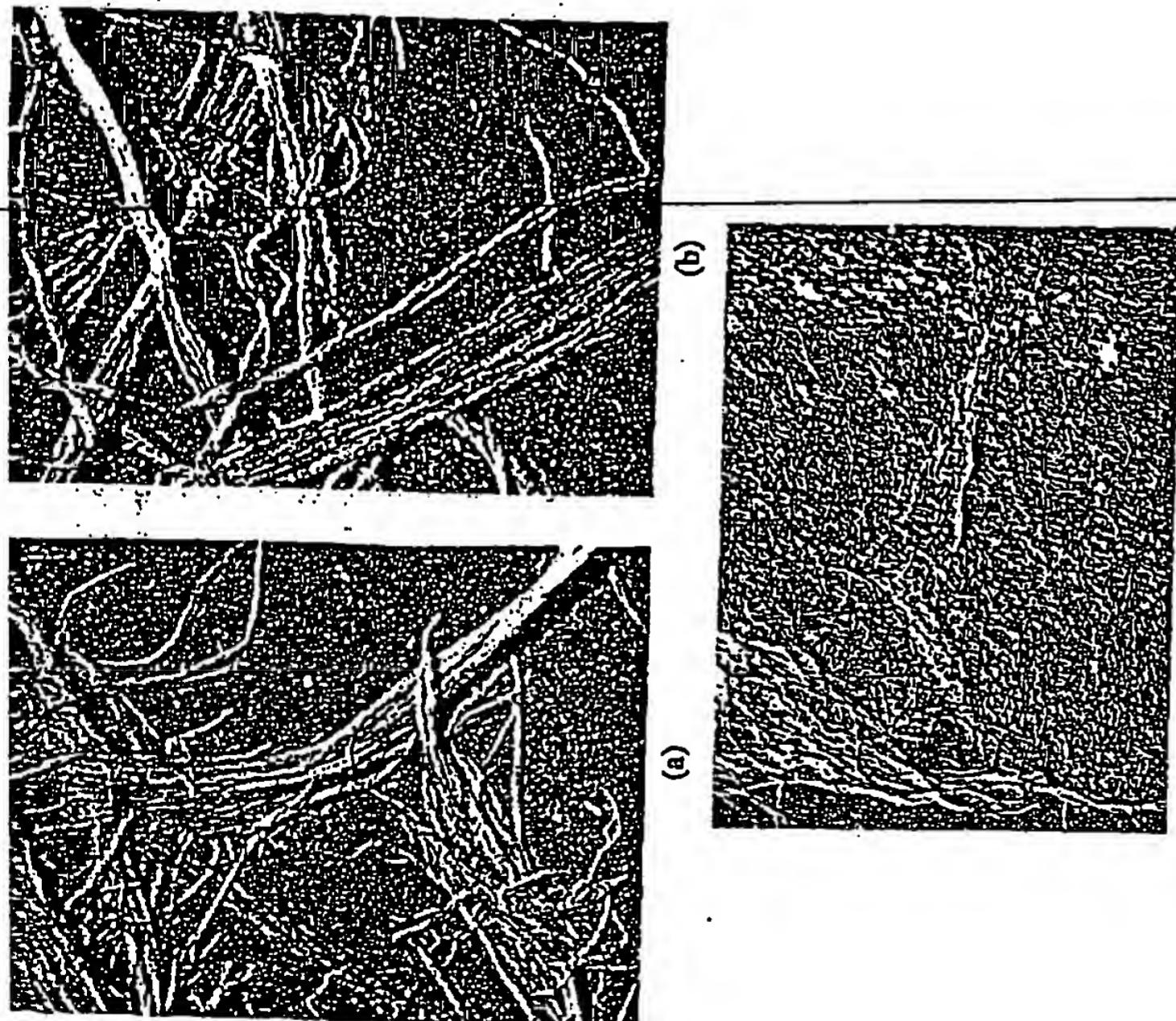


Fig. 4.6. Electron micrographs of cellulose microfibrils and elementary fibrils, from (a) purified sprucewood pulp and (b) cotton, disintegrated by ultrasonic irradiation of the fibers in water suspension (x 27,000) (Ranby) (c) As occurring in the primary wall of eucalypt fibers (x 9,000) (Wardrop)

other to an extent which permits an evaluation of how far these fibrils constitute structural entities, Figure 4.6 (635). Although the values of the average width of the elementary fibrils have tended to decrease with improved resolution of the electron microscope, it is now fairly well established, that they are about 100 Å wide, 30 Å thick and of infinite length. The elementary fibrils of wood cellulose may be less wide than those of cotton, whereas animal and algal cellulose elementary fibrils may be a little wider, possibly accounting for the differences noted in the corresponding x-ray diagrams. These structural units are often intimately aggregated to microfibrils about 200–250 Å wide by means of hydrogen bonding between the respective surface layers, Figure 4.7 (235). On hydrolysis, the elementary fibrils first tend to aggregate still further (812) and then eventually disintegrate into short fragments, *micelles*, of the

attack. The degradation of the more crystalline regions first occurs in the vicinity of the amorphous regions. After the initial phase of the hydrolysis, which removes the amorphous matter and degrades the crystalline portion to a more or less constant level of DP 100–200, a second phase follows, in

by nitrogen bonding (a modification suggests hydrogen bonding between the $a-b$ planes in the 101 direction (234)). The chains of adjacent $a-b$ planes run in opposite directions and are staggered as to their vertical arrangement by the length of half a monomer. Figures 4.10-12 show the

last for wood cellulose as for cotton. During that phase, the micellar length and DP of the residue from the latter material remains almost constant, whereas that from wood cellulose gradually decreases in micellar length and DP (559, 582). This indicates a higher degree of crystalline perfection in the cotton cellulose micelles (378, cf. 719). Not only alkali treatment but also drying of the original cellulose induces changes in the course of subsequent hydrolytic degradation and results in a lower DP level (378, 502), thus reflecting changes in the super-structure of cellulose. Mechanical influence, such as crushing or bending of the fibers, which is known to introduce slip planes and irregularities in the association of the elementary fibrils to larger structures, cf. Chapter 3, may cause similar disturbance within the fibrils and lead to phenomena of the same type as those previously mentioned for alkaline swelling or drying.

The detailed arrangement of the crystalline regions has been deduced from x-ray data (34, 265, 325, 550, 629, 739). An elementary cell with a rhombic symmetry was originally proposed (629), later modified (550) to a monoclinic cell with the dimensions:

$$\begin{array}{ll} a = 8.3 \text{ \AA} \\ b = 10.3 \text{ \AA} \\ c = 7.9 \text{ \AA} \\ \beta = 84^\circ \end{array}$$

as illustrated in Figure 4.9 (550). The cellulose chains are arranged parallel to the b axis and have the symmetry of a diagonal screw axis (a modification suggests a distortion of every second monomer (83, 590)). The chains are parallel to each other and are considered to hold together in the $a-b$ plane.

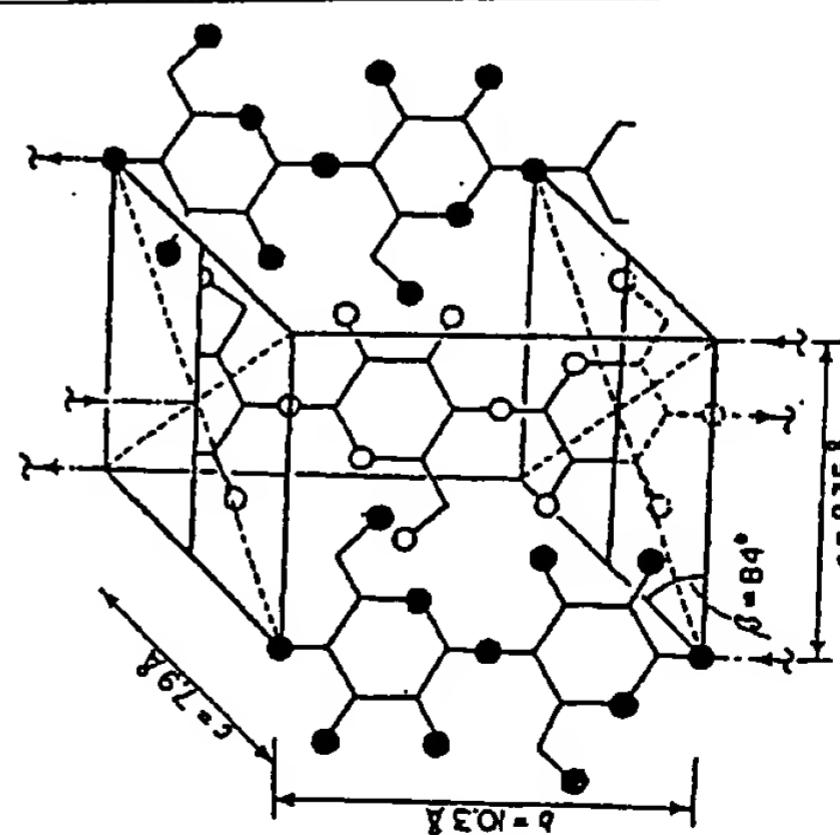


Fig. 4.9. The monoclinic elementary cell of cellulose (Meyer-Misch)

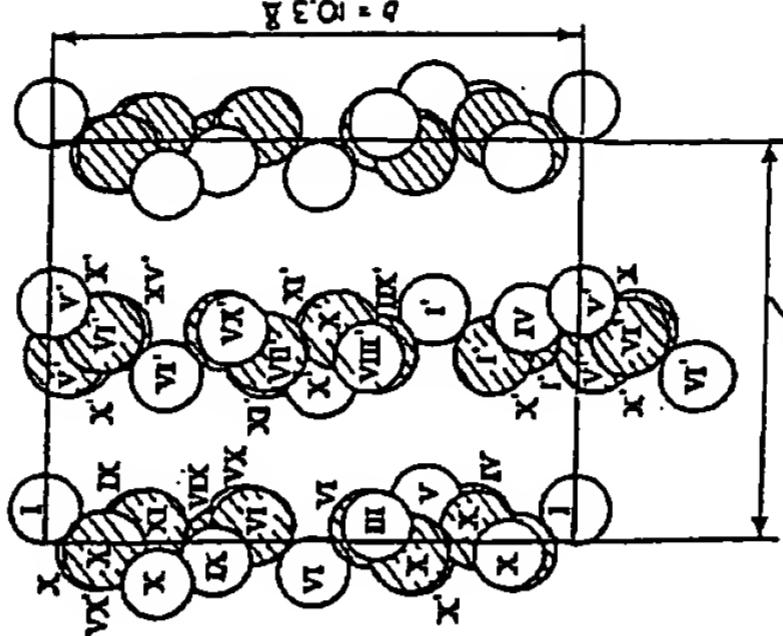
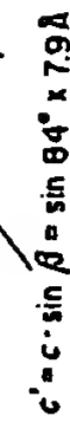


Fig. 4.11. Projection of the elementary cell of cellulose on the $b-c$ plane perpendicular to the a -axis (Wisc-Jahn).



detailed arrangement of the atoms of the elementary cell in the three projections. The hydroxyl oxygen of two adjacent chains are at a distance of only 2.5 Å in the direction of the a axis, allowing complete hydrogen bonding. The closest distance between the atoms belonging to different $a-b$ planes is about 3.1 Å (cf. however (2.34)), allowing only weaker forces, i.e. the attraction of the OH-dipoles and the permanent electric moment

ounds (cuam or cuen solutions) or unstable substitution compounds, predominantly cellulose xanthate. Its x-ray diffraction diagram is shown in Figure 4.13 (635) in comparison to that of cellulose I. The x-ray studies have revealed (34, 114, 550) that cellulose II has a monoclinic elementary cell of the dimensions:

$$\begin{aligned}a &= 8.1 \text{ \AA} \\b &= 10.3 \text{ \AA} \\c &= 9.1 \text{ \AA} \\&\beta = 62^\circ\end{aligned}$$

which contains four glucose monomers as in the case of cellulose I. The spatial arrangement of the chains is shown in Figure 4.14 in comparison with cellulose I. It is seen that the transformation involves a slight dis-

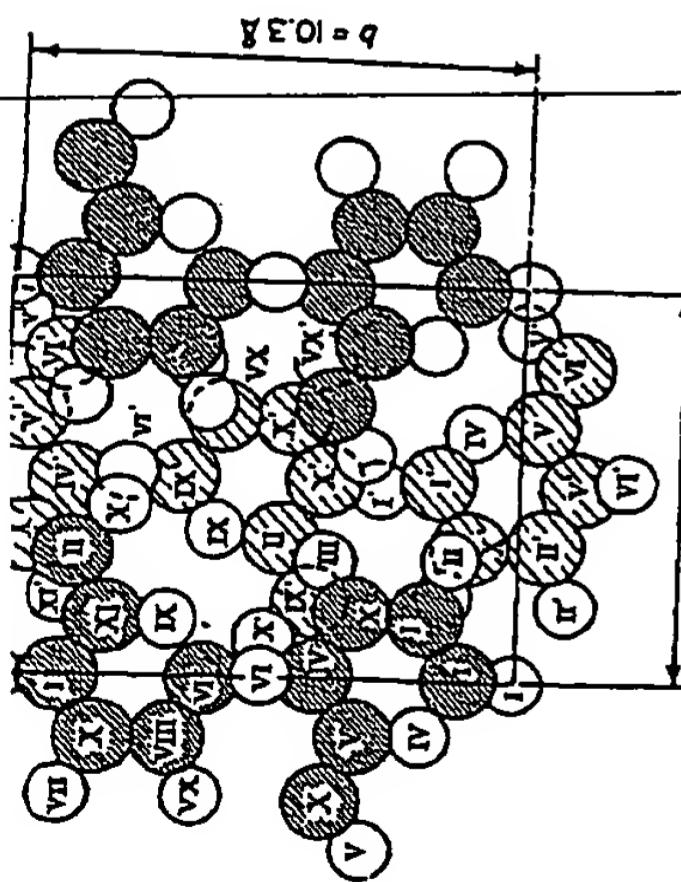


Fig. 4.12. Projection of the elementary cell of cellulose on the $a-b$ plane perpendicular to the c -axis (Wise-Jahn)

of the C-O-C groups. The strongest forces, of covalent bonds, operate in the direction of the b axis. Recent work (83, 590, 618) indicates, that the glucose units have the C1 *conformation* (hair form) and every second unit slightly twisted (20-30°). Infrared analysis further suggests, that all hydroxyls are engaged in hydrogen bonding, and that an *intrachain* hydrogen bond is present between the 3-hydroxyl and the ring oxygen of the adjacent monomer (488, 524, 797). If that bond prevails also after mercerization, it would probably decrease the reactivity of the 3-hydroxyl in substitution reactions (143), cf. the subsequent section. These recent observations cause only relatively small adjustments of the classical model. A more radically new concept has also been advanced for discussion (787). Work on other polymers, such as polyethylene, nylon, etc., has indicated that chain *folding* can occur to give crystals of uniform thickness (197, 430), and it was suggested (787) that similar folding of cellulose chain molecules in the 101 plane occurs, giving discontinuities for every 500 \AA. The comparative rigidity of the cellulose molecules in solution (e.g. 319) is an argument against such a super-structure, but investigations on steric models indicate a U-turn diameter of only about 10 \AA (644), and cellulose derivatives and even cellulose have been obtained from very dilute solutions in the form of regular, compact, lamellar crystals of microscopic size, similar to the single crystals of linear synthetic polymers mentioned above (644).

The crystalline structure described is valid for native cellulose. There are several polymorphous forms, of which that of *regenerated cellulose* is the most important. This form is also designated *cellulose II* in contrast to *cellulose I*, native cellulose. Cellulose II is formed upon regeneration of cellulose from its solid addition compounds, such as the acid, water or alkali celluloses (cf. below), as well as from solutions of addition com-

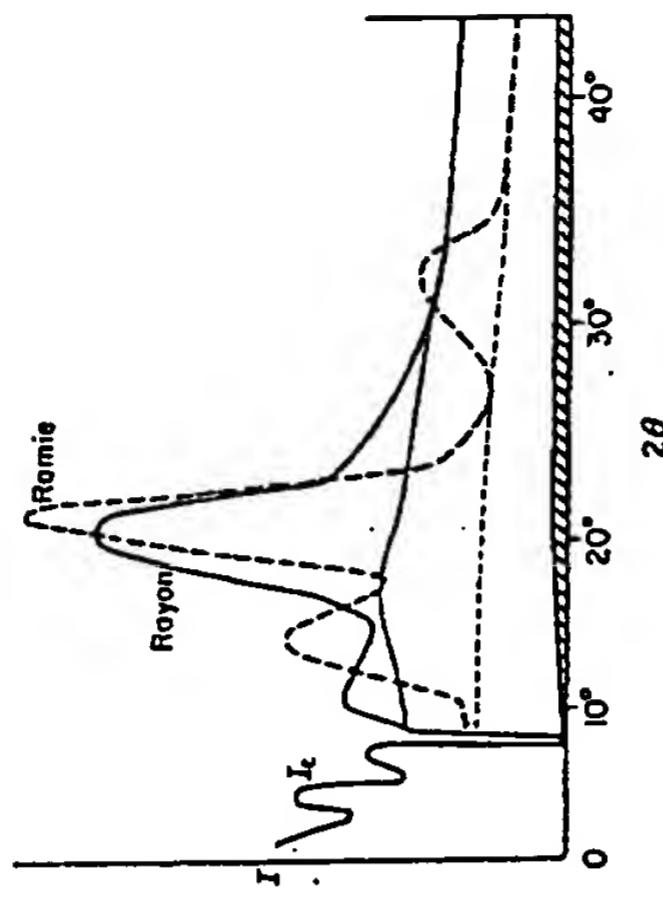


Fig. 4.13. X-ray scattering diagrams from cellulose I (ramie) and cellulose II (rayon). I_c is a reflection from a standard sample (Hermans)

tortion of the chains out of the $a-b$ plane to form new hydrogen bonds in the 101 direction. It is thereby noticed that the chains thus interconnected run in an opposite direction to the situation in cellulose I. Cellulose II seems to be the thermodynamically more stable form. On heating cellulose II to high temperatures in glycerol or alkali, it is converted to a new crystalline form which closely resembles cellulose I (68, 139, 326, 459, 551), but is probably a separate type, called high-temperature cellulose or cellulose IV (327, 372, 416), with an orthorhombic symmetry and the approximate dimensions:

$$\begin{aligned}a &= 8.1 \text{ \AA} \\b &= 10.3 \text{ \AA} \\c &= 7.9 \text{ \AA} \\&\beta = 90^\circ\end{aligned}$$

indicating a denser packing than both cellulose I and II (corresponding to a density of 1.62 as compared to 1.59 for cellulose I and II and about 1.50

normally used in pulping processes and is therefore of little importance, as is cellulose III, observed to form on the decomposition of ammonia cellulose (326).

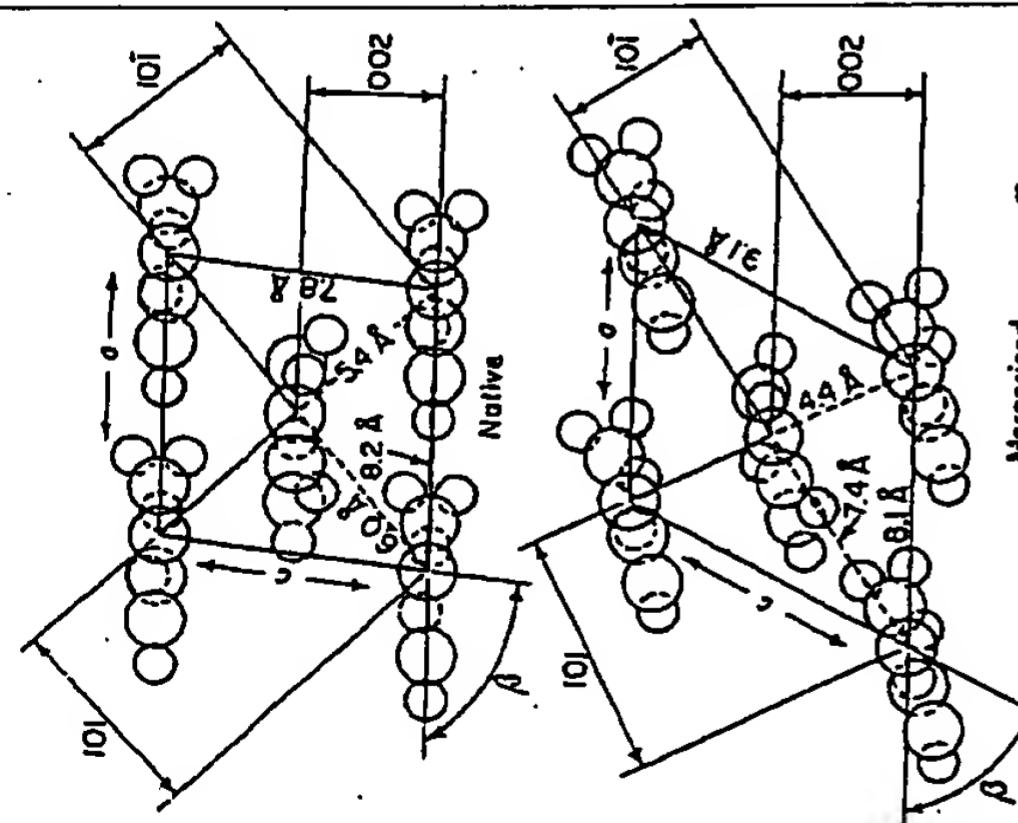


Fig. 4.14. Cross-sections of the unit cells of cellulose I and II lattices, cut at right angles to the b -axis, i.e. at right angles to the cellulose chains

The importance of the super-molecular structure of cellulose for its properties and its heterogeneous reactions is obvious. Only a limited fraction of its hydroxyl groups is available for interaction with water, and cellulose hence remains insoluble in water in spite of its polarity. Likewise most of its hydroxyl and acetal groups are comparatively inaccessible to chemical reagents unless the super-structure is influenced by treatment with strong swelling agents. The physical properties are influenced not only by the average molecular length and the length distribution, but also by the degree of crystallinity.

4. REACTIVITY

As just indicated, the influence of the super-molecular structure on the reactivity of cellulose is profound and has to be considered in all cellulose reactions of heterogeneous type. However, obviously the various possibilities of cellulose reactions are determined by its molecular constitution. Like all carbohydrates, the cellulose molecule is capable of reactions at its hydroxyl and acetal groups, as well as at the aldehydic end groups. The

reactions now described for cellulose will be principally possible also for the hemicelluloses, and in the section on hemicelluloses therefore only those deviations will be pointed out which result from the different carbohydrate structure of those compounds.

The hydroxyl groups react with *addition*, *substitution* and *oxidation* agents and the acetal groups undergo *hydrolysis* in acid as well as in alkaline medium. The aldehydic end groups can be *reduced* to alcohol groups, *oxidized* to carboxyl groups or *rearranged* under the influence of alkali to form either alcohol or carboxyl end groups. Since the redox and rearrangement reactions all influence the ease of hydrolysis of the glucosidic bonds, they will be treated together with hydrolysis under the heading of chemical *degradation reactions*. This category of reactions include those of importance for the pulping processes, whereas the addition and substitution reactions are of interest mainly in connection with the use of dissolving pulps. A special variant of the substitution reactions, called *grafting*, involves the incorporation of copolymers, e.g. of the polyvinyl type, usually by reacting cellulose with the monomer in a redox system, such as ceric ions (379, 710a). A radical mechanism is involved. One addition reaction, that of carbohydrates and water, is of predominant importance in the use of paper pulps. To facilitate the understanding of the various reactions, the analogous reactions with low-molecular compounds will first be treated, followed by a discussion of the reaction of cellulose in homogeneous solution, and finally the corresponding reaction in the heterogeneous two-phase system will be described.

A. Degradation reactions

(a) *Degradation of low-molecular model compounds*. Hydrolysis of glucosidic bonds occurs in both acid and alkaline medium, although much faster at low pH. The rate of reaction also varies with the type of sugar and glycone. Table 4.18 (161, 831) gives the relative rates of hydrolysis of various methyl glycopyranosides, showing the relative stability of glucosides in acid medium and of mannosides in alkaline medium. Glycosiduransides are much more easily hydrolyzed in acid medium than

Table 4.18. Relative rates of hydrolysis of various methyl glycopyranosides (161, 831)

	Acid hydrolysis	Alkaline hydrolysis
Methyl- α -glucoside	1.0	1.0
Methyl- β -glucoside	1.9	10.0
Methyl- α -mannoside	2.4	11.0
Methyl- β -mannoside	5.7	4.4
Methyl- α -galactoside	5.2	3.9
Methyl- β -galactoside	9.3	23.0
Methyl- α -xyloside	4.5	4.8
Methyl- β -xyloside	9.0	23.0

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WOOD CHEMISTRY

Fundamentals and Applications

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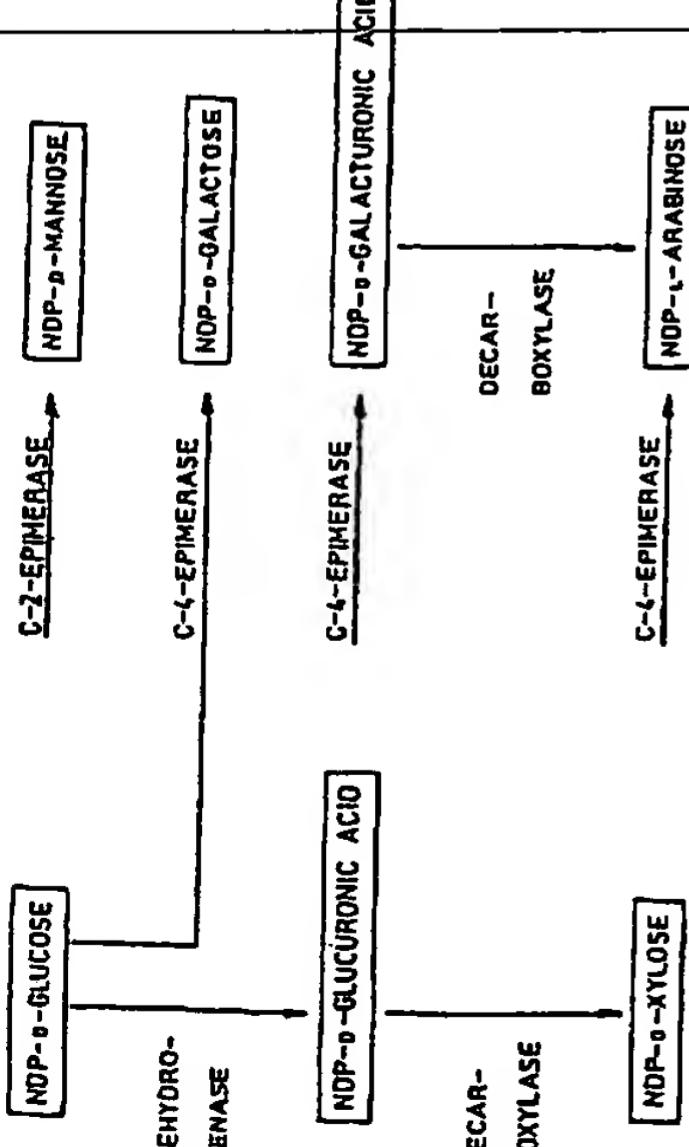


Fig. 3-4. Simplified representation of the formation of hemicellulose precursors from UDP-D-glucose or GDP-D-glucose. Note that NDP (nucleotide diphosphate) means either UDP or GDP.

3.2.1 Molecular Structure

Although the chemical structure of cellulose is understood in detail, its supermolecular state, including its crystalline and fibrillar structure is still open to debate. Examples of incompletely solved problem areas are the exact molecular weight and polydispersity of native cellulose and the dimensions of the microfibrils.

Cellulose is a homopolysaccharide composed of β -D-glucopyranose units which are linked together by $(1 \rightarrow 4)$ -glycosidic bonds (Fig. 3-5). Cellulose molecules are completely linear and have a strong tendency to form intra- and intermolecular hydrogen bonds. Bundles of cellulose molecules are thus aggregated together in the form of microfibrils, in which highly ordered (crystalline) regions alternate with less ordered (amorphous) regions. Microfibrils build up fibrils and finally cellulose fibers. As a consequence of its fibrous structure and strong hydrogen bonds cellulose has a high tensile strength and is insoluble in most solvents. The physical and chemical behavior of cellulose differs completely from that of starch, which clearly demonstrates the unique influence of stereochemical characteristics. Like cellulose, the amylose component of starch consists of $(1 \rightarrow 4)$ -linked D-glucopyranose units, but in starch these units are α -anomers. Amylose

3.2 Cellulose 53

occurs as a helix in its solid state and sometimes also in solution. Amylopectin, the other starch component, is also a $(1 \rightarrow 4)$ - α -glucan but is highly branched. The branched structure accounts for its extensive solubility, since no aggregation can take place.

The crystalline structure of cellulose has been characterized by X-ray diffraction analysis and by methods based on the absorption of polarized infrared radiation. The unit cell of native cellulose (cellulose I) consists of four glucose residues (Figs. 3-6 and 3-7). In the chain direction (c), the repeating unit is a cellobiose residue (1.03 nm), and every glucose residue is accordingly displaced 180° with respect to its neighbors, giving cellulose a 2-fold axis. It has now been established and largely accepted that all chains in native cellulose microfibrils are oriented in the same direction, that is, they are parallel (Fig. 3-7). There are two hydrogen bonds within each cellulose chain, namely from O(6) in one glucose residue to O(2)H in the adjacent glucose and also from O(3)H to the ring oxygen, as shown in Fig. 3-8. The chains form a layer in the a-c crystallographic plane, where they are held together by hydrogen bonds from O(3) in one chain to O(6)H in the other. There are no hydrogen bonds in cellulose I between these layers, only weak van der Waal's forces in the direction of the b-axis. Native cellulose therefore has a chain lattice and a layer lattice at the same time.

Regenerated cellulose (cellulose II) (Fig. 3-6) has antiparallel chains (Fig. 3-9). The hydrogen bonds within the chains and between the chains in the

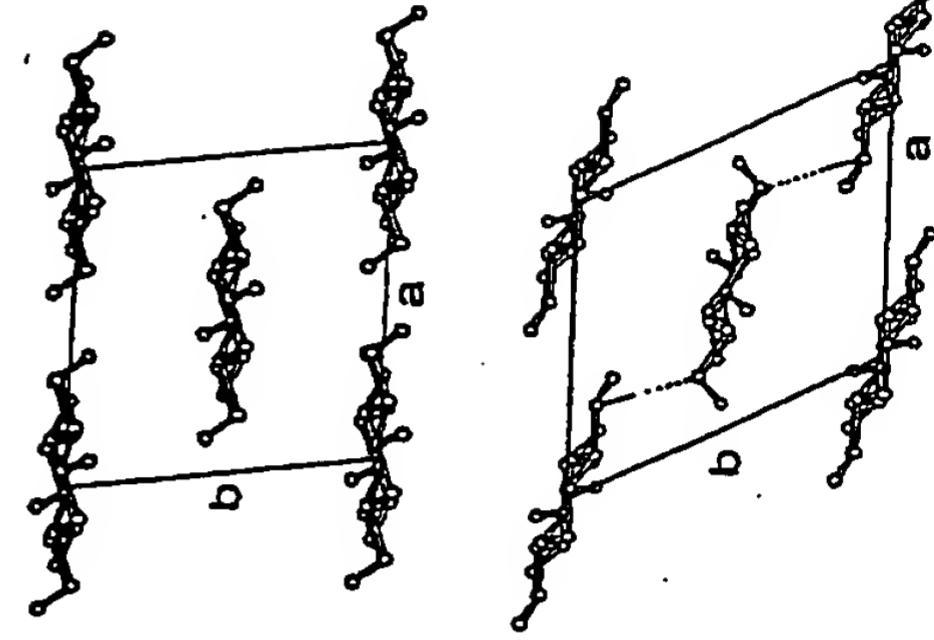


Fig. 3-6. Axial projections of the structures of native cellulose (cellulose I, above) and regenerated cellulose (cellulose II, below). (Reproduced from Kolpak et al., 1978, Polymer 19, 123-131, by permission of the publishers, IPC Business Press Ltd. ©.)



Fig. 3-5. Structure of cellulose. Note that the β -D-glucopyranose chain units are in chair conformation (C_1), and the substituents HO-2, HO-3, and CH_3OH are oriented equatorially.

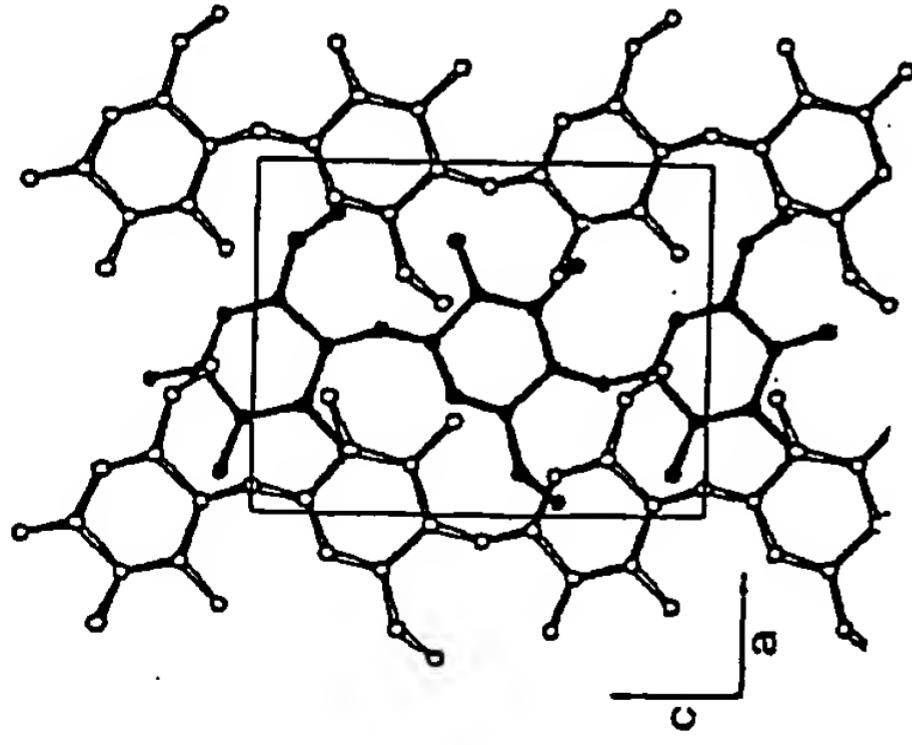


Fig. 3-7. Projection of the chains in cellulose I perpendicular to the ac plane. The center chain (black) is staggered but is parallel with the two corner chains (Gardner and Blackwell, 1974).

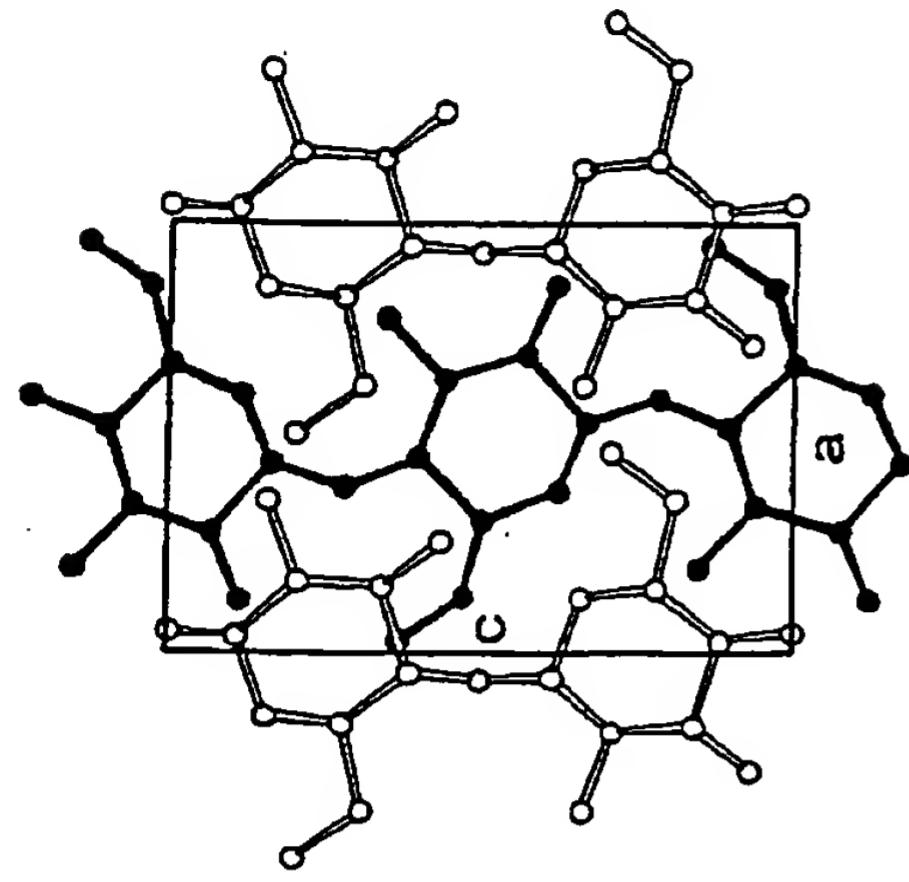


Fig. 3-9. Projection of the chains in cellulose II perpendicular to the ac plane. The center chain (black) is staggered and antiparallel to the corner chains. (Reproduced from Kolpak et al., 1978, *Polymer* 19, 123-131, by permission of the publishers, IPC Business Press Ltd. ©.)

$a-c$ plane are the same as in cellulose I. In addition, there are two hydrogen bonds between a corner chain and a center chain (Fig. 3-6), namely from $O(2)$ in one chain to $O(2)H$ in the other and also from $O(3)H$ to $O(6)$. Cellulose II is formed whenever the lattice of cellulose I is destroyed, for example on swelling with strong alkali or on dissolution of cellulose. Since the strongly hydrogen bonded cellulose II is thermodynamically more stable than cellulose I, it cannot be reconverted into the latter. All naturally occurring cellulose has the structure of cellulose I. Celluloses III and IV are produced when celluloses I and II are subjected to certain chemical treatments and heating.

The proportions of ordered and disordered regions of cellulose vary considerably depending on the origin of the sample (cf. Table 9-1). Cotton cellulose is more crystalline than cellulose in wood.

3.2.2 The Chain Length and Polydispersity of Cellulose

The polymer properties of cellulose are usually studied in solution, using solvents, such as CED or Cadoxen (see Section 9.2). On the basis of the solution properties, conclusions can be drawn concerning the average molecular weight, polydispersity, and chain configuration. However, the

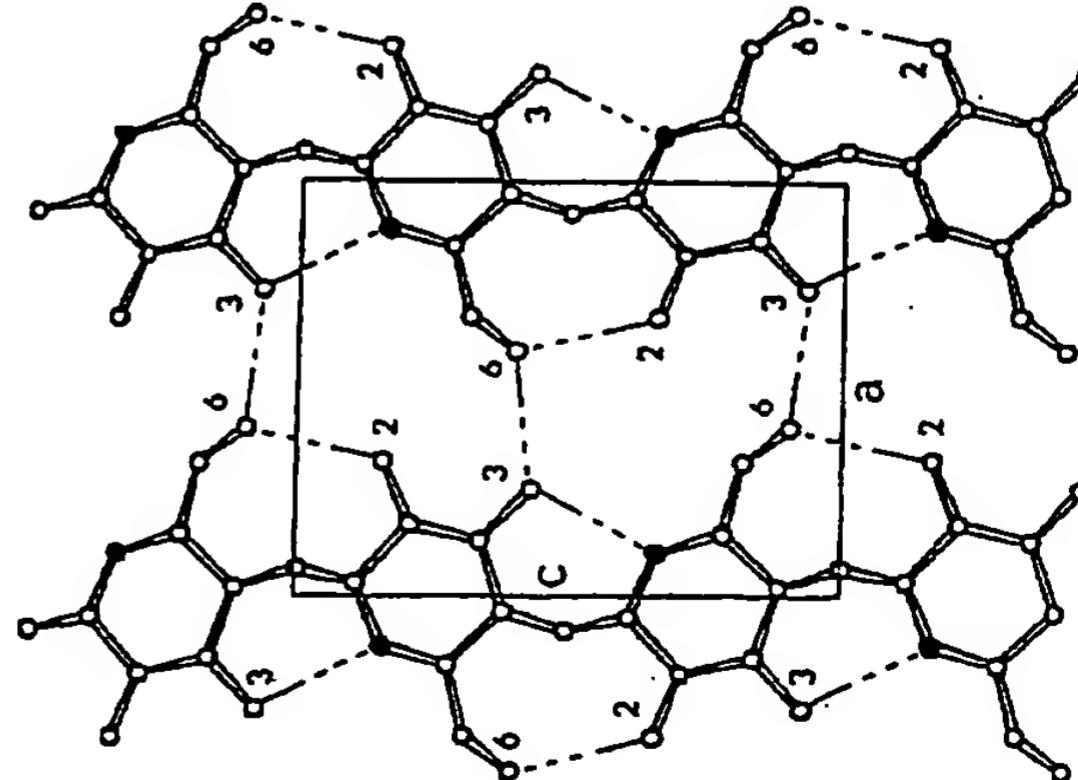


Fig. 3-8. Projection of the (O2O) plane in cellulose I, showing the hydrogen bonding network and the numbering of the atoms. Each glucose residue forms two intramolecular hydrogen bonds ($O_3-H\cdots O_{5'}$ and $O_6-H\cdots O_3'$) and one intermolecular bond ($O_6-H\cdots O_3$). (Slightly modified from Gardner and Blackwell, 1974.)

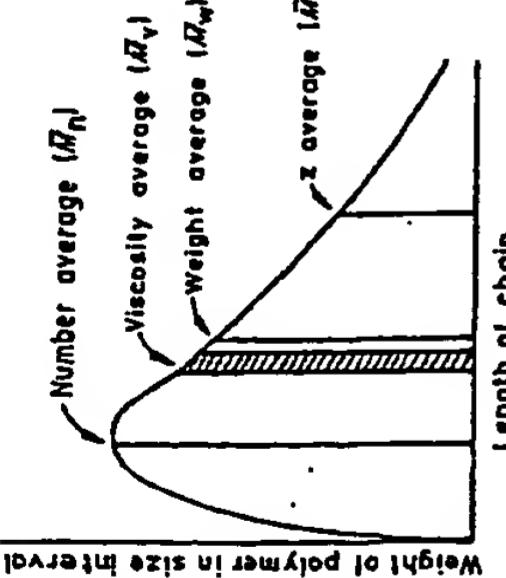


Fig. 3-10. The molecular weight distribution and the average molecular weights of a typical polymer (Billmeyer, 1965).

isolation of cellulose from wood involves risk for some degradation resulting in a reduced molecular weight.

The distribution of molecular weights can be presented statistically as illustrated by Fig. 3-10 where the weight of polymer of a given size is plotted against the chain length. The experimental measurements give an average value of the molecular weight and some methods also a molecular weight distribution. For any polydisperse system, these average values differ from each other depending on the method used. The number average molecular weight \bar{M}_n * can be measured using osmometry or by determining the number of reducing end groups. The weight average molecular weight \bar{M}_w can be deduced from light scattering data. Sedimentation equilibrium data attainable by ultracentrifugation technique give so-called \bar{M}_z values. Finally, \bar{M}_v refers to the molecular weight calculated on the basis of viscosity measurements. For cellulose, the relationship between molecular weight and degree of polymerization (DP) is $DP = M/162$, where 162 is the molecular weight of anhydroglucoside unit. The ratio \bar{M}_w/\bar{M}_n is a measure of polydispersity corresponding to the width of the molecular weight distribution and ranges for typical polymers from 1.5-2.0 to 20-50.

Molecular weight measurements have shown that cotton cellulose in its native state consists of about 15,000 and wood cellulose of about 10,000 glucose residues. Some polydispersity data on cellulose derivatives and polysaccharides are shown in Table 3-1. There are indications that the native cellulose present in the secondary cell wall of plants is monodisperse, that is, contains only molecules of one size. In such a case, number and weight average molecular weights ought to be identical. The cellulose in the primary cell wall, on the other hand, which has a lower average molecular

TABLE 3-1. Polydispersity Values (\bar{M}_w/\bar{M}_n) of Different Polysaccharides^a

Macromolecule	Source	$\bar{M}_w \times 10^{-4}$	\bar{M}_w/\bar{M}_n
Cellulose nitrate	Birch	27 ^b	1.9 ^c
Cellulose nitrate	Ramie	24 ^b	1.7 ^c
Amylose	Potato	8.8 ^c	1.9 ^d
Xylan	Birch	0.8 ^e	2.3 ^e
Xylan	Elm	0.7 ^e	2.4 ^e
Amylopectin	Waxy corn	1700 ^b	116 ^f
Hydrolyzed amylopectin	Waxy corn	15 ^b	25 ^f
Glycogen	Sweet corn	190 ^b	15 ^f
Hydrolyzed glycogen	Sweet corn	20 ^b	6.3 ^f
Glycogen	Rabbit liver	390 ^b	6.6 ^f

^a From Gorling (1962).

^b \bar{M}_w by light scattering.

^c \bar{M}_n by viscometry from $[\eta] = 0.0091$ DP.

^d Calculated from fractionation data.

^e \bar{M}_n by osmometry.

^f \bar{M}_w from the alkali number.

* \bar{M}_w from sedimentation and diffusion used instead of \bar{M}_n ; usually $\bar{M}_w > \bar{M}_w > \bar{M}_n$.

weight, is evidently polydisperse, being similar in this respect to the hemicelluloses.

3.2.3 The Configuration of Cellulose Molecules

Based on properties in solution such as intrinsic viscosity and sedimentation and diffusion rates, conclusions can be drawn concerning the polymer configuration. Like most of the synthetic polymers, such as polystyrene, cellulose in solution belongs to a group of linear, randomly coiling polymers. This means that the molecules have no preferred structure in solution in contrast to amylose and some protein molecules which can adopt helical conformations. Cellulose differs distinctly from synthetic polymers and from lignin in some of its polymer properties. Typical of its solutions are the comparatively high viscosities and low sedimentation and diffusion coefficients (Tables 3-2 and 3-3).

Any linear polymer molecule, even a reasonably stiff rod, will coil randomly, provided the chain is sufficiently long. In addition to the size of the monomer units, the tendency for coiling is affected by the forces between the units as well as the interaction between the polymer and the solvent. One measure of the stiffness of a polymer molecule is the end-to-end distance (R). For a polydisperse polymer the root-mean-square average of R (\bar{R}^2)^{1/2} is used. R is affected by the properties of the polymer itself as well as by the interaction of the solvent. The better the solvent the more the polymer

*The SI system (Système International d'Unités) recommends the term *relative molecular mass* instead of *molecular weight*, but because the SI term is not yet universally adopted in the polymer chemistry the latter term is used throughout this book.

RELATED PROCEEDINGS APPENDIX

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